Humidity-Responsive Tripeptide Crystals

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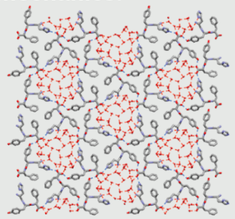
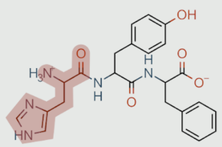
Introduction

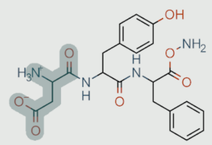
Water-responsive (WR) materials convert changes in relative humidity (RH), to mechanical work, with potential applications in energy storage and soft robotics.[1,2] Despite these promising applications, mechanisms underpinning the behavior are poorly understood. Self-assembled tripeptides are simple, tunable materials which provide useful systems to probe mechanics.[3,4]



HYF crystal structure



HYF
water-responsive



DYF
less water-responsive

Experimental Methods

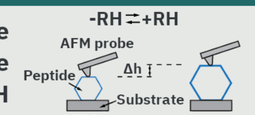
To understand the change in peptide structure with RH, both structures were investigated between 10% and 90% RH using various techniques:

A customized atomic force microscope (AFM) was used to quantify shape changes in HYF/DYF upon hydration. This was used to measure actuation force.

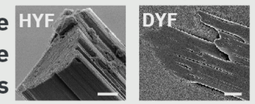
Powder X-ray diffraction (PXRD) was conducted to observe lattice deformation.

Change in mass was investigated using dynamic vapor sorption (DVS).

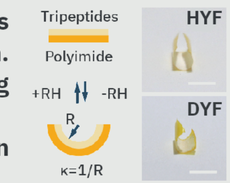
Changes in supramolecular configuration were followed using FTIR.



AFM microscopy



Crystal SEM images



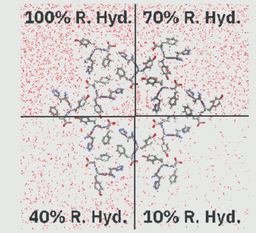
Polyimide film deformation

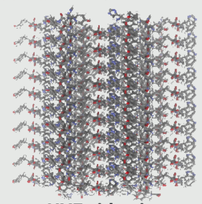
Computational Methods

Molecular dynamics (MD) simulations were carried out of HYF and DYF pores at different relative hydration (R. Hyd.) levels (% of water relative to standard density) using the CHARMM forcefield.[5] Input structures were generated from single crystal X-ray diffraction data.

Average pore width and pore solvent occupancy (the mean number of water molecules in the pore over a simulation) were measured for each R. Hyd. level.

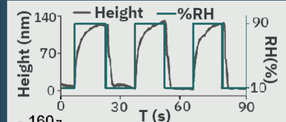
Simulations were run in triplicate.

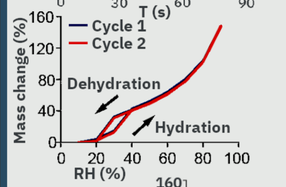




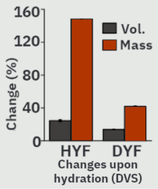
HYF side view

Key Results





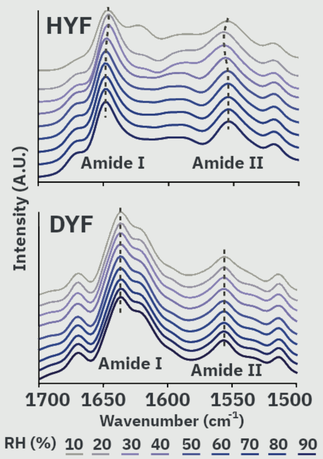
Video content

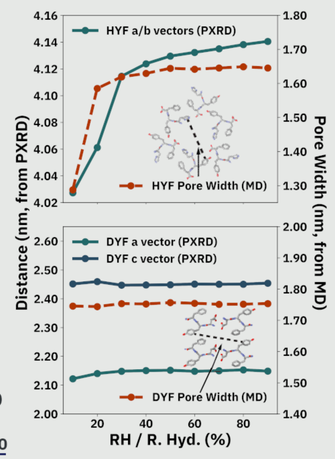


Results from experiment and simulation show the nonlinear actuation of HYF at around 20-30% RH. Changes in simulation pore size are in agreement with PXRD data (far right), showing the increased water-responsiveness of HYF *vis a vis* DYF.

FTIR data, showed shifts in the Amide I/II bands for HYF upon dehydration (suggesting stronger H-bonds between peptides),[6] DYF remained unchanged (right).

DVS showed substantially more water uptake than expected from AFM data (left), suggesting water adsorption at the next hierarchical level (crystal surfaces, interfaces), evidenced by the hysteresis loop between 20% and 40% RH during hydration/ dehydration cycles.





Conclusions

Maximum energy density of a WR material is primarily determined by the total number of absorbed/desorbed water molecules and their chemical potential.

To efficiently convert that potential to mechanical energy, the H-bond network should be strong enough to transfer pressure to surrounding supramolecular structures. These hierarchical organizations can provide high mechanical stiffness and ductility to effectively store energy elastically, or transfer it to external loads.

Our findings provide insights into a general description of WR behaviour, and could serve as design criteria for synthetic development of high energy WR actuators.

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